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REPORT Item 0002

for the contract

Investigation of K-Mixing in Nuclei with Isomeric States

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Submitted by
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I. Introduction

Nuclear isomers represent a unique opportunity to store high energy densities for long times. The question is how can one control the release of this energy at time scales of nano- or microseconds. Many applications would take benefit of such high energy density release (without implying a nuclear process) even in a non-coherent way.

As the nucleus is the smallest structural unit of the matter, according to the quantum mechanics laws, the constituents of the nucleus (the nucleons) will have the highest possible circulation velocities. These particles in motion can absorb electromagnetic radiation determining a transition of the nucleus in an excited state. Usually, the nucleus decays from this excited state in very short times ($<10^{-18}$ sec.) and reradiate the absorbed energy. The case of nuclear isomers is of particular interest since in such states there are selection rules which inhibit the coupling of the particle motion to the electromagnetic field resulting in a retardation of the reradiation process. Such states can store the energy for hundreds and thousands of years.

For long time it was thought that it is impossible to trigger the release of the energy stored in this states. Recently, it was found that nuclei in the A~180 mass region are characterized by the existence of a giant pumping resonance located at an energy near

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2.5 MeV [1]. This giant resonance acts as a gateway through which the selection rules making an isomer long-lived could be violated. If an isomeric level initially stored an energy of 2.0 MeV, only 0.5 MeV would be needed to reach the gateway at 2.5 MeV. The absorption of an x-ray photon of that energy would excite the system to such a level which would be very strongly coupled to the electromagnetic fields. The sum of the stored energy and that of the trigger x-ray photon would then be promptly emitted, or dumped, as gamma rays. Nuclei as Lu, Hf and Ta are characterized by the existence of 4 and 5 quasipaeticle isomeric states which are distiguished by the combination of high energies and long half-lives. Such states are termed K-isomers because the long lifetimes of the spontaneous radiative decay are attributed to structural changes forbiden by K-quantum numbers. In this mass region the nuclei are deformed and the projection of the total angular momentum upon the symmetry axis contributes a quantum number, K, which can change during a radiative transition by no more than the multipolarity of the mediating moment. Transitions from the high-K isomer to the rotational states of a low-K band are forbidden, and so long lifetimes are inevitable. One of the most promissing nucleus is the ^{178}Hf. This nucleus has an isomeric state with angular momentum $I=16\hbar$ ($K=16$) located at 2.45 MeV with a half-life of 31 year.

II. Previous Results

Proposals to trigger the release o the energy of a nuclear isomer by exciting it to some higher level associated with freely radiating states have been known for over a decade [2]. Such schemes require the existence at an energy near that of the isomer of a state of mixed-K which could be reached by an allowed electromagnetic transition of low multipolarity from both high and low K states. Such K-mixing states have been reported in ^{180}Ta [3] and described in ^{174}Hf [4]. In the case of ^{180}Ta the resonant absorption of x-rays exited the nucleus from the 2 quasiparticle isomeric state to a K-mixing level at 2.8 MeV through an integrated cross section of 1.2 x 10^{-25} $cm^2 \cdot keV$ from which cascading to the ground state was found to occur [5].

Several methods to trigger the energy release from the nuclear isomers were proposed during the last decade. One of them consist in the inelastic scattering of alpha particles on the isomeric target. Following this idea two such measurements (in 1996 and 1997) were performed at the Orsay Tandem accelerator. The experiments consisted in the irradiation of the target containing isomeric nuclei of ^{178m2}Hf with alpha particle and the detection of the emitted gamma rays with the Crystall Ball in coincidence with the backscattered alpha' particles detected with an annular Si detector. The results from 1996 [6] strongly suggested the possibility to get the triggering of the energy stored in the ^{178m2}Hf isomer. In 1997 the experiment was repeated using a better charged particle detector. Several members of our group (K1) supported this measurement and were invited to participate at the experiment. At that time we managed to get only screen dumps of the spectra acquired on-line during the measurement. We have analyzed the digitized screen dumps and reported the results in the Report to the Item 0001 of the present contract [7]. Unfortunately, the French group refused to share the raw data with the other participants to the experiment and we could not benefit of the entire statistics got during the measurement. However, despite the fact that the French group claim that the experiment didn't work right, the results we obtained from the analysis of the digitized screen dumps were similar to the ones obtained from the experiment performed in 1996. The most important conclusions we got from these data were: the possibility to trigger the energy release from the isomeric state and that the energy needed for this process is lower that the energy resolution of the charged

particle detector, namely ~125 keV (in agreement with the results of the systematics on the position of the gateway states in the A~180 mass region, as mentioned earlier).

This kind of methods to get the triggering effect are not practical for application purposes since they require huge accelerators to produce intense particle beams. Also, the target production needs complicated operations. More appropriate seem to be the (gamma, gamma') processes consisting of the resonant absorption of a photon by a nucleus in the isomeric state causing the passage in another excited state which freely decays to the ground state. Encouraged by the results obtained from the (alpha, alpha') measurements we performed a (gamma, gamma') experiment at the Center for Quantum Electronics, University of Texas at Dallas using a low energy x-ray device.

III. Experimental Details

A sealed plastic target containing 6.3 x 10^{14} isomeric nuclei in a 1 cm diameter well was exposed to the brehmsstrahlung radiation from a dental x-ray unit operating at 15 mA. The voltage applied to the anode of the x-ray tube was a half-wave rectification of 60 Hz ac. The endpoint varied during a half period up to the nominal maximum of 70 or 90 kV, as selected. Better signal-to-noise, S/N resulted from use of the 90kV. The counting circuit was enabled for only the peak 48% portion of each radiation pulse so as to maximize S/N.

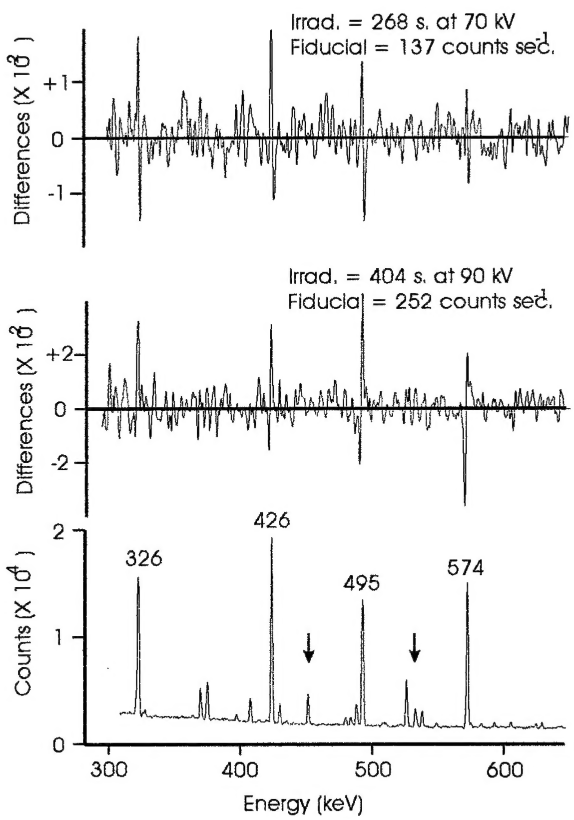

Figure 1. Spectra of the gamma photons emitted by the isomeric target. Each of the upper and middle panels show differences between data taken with and without x-rays for the particular conditions marked on the figure. The parameters of the irradiations are summarized by the corresponding total durations and endpoints; while the different geometric efficiencies are proportional to the counting rates reported in the fiducial line from the spontaneous decay of ^{172}Lu at 1093.7 keV. A typical comparison spectrum is shown in the bottom panel. Major lines from the spontaneous decay of the ^{178}Hf isomer are labeled and minor lines are marked with arrows. Lines not marked belong to impurities. The effects of a slow drift of less than a channel-width in the energy calibration are seen in the apparent differentiation of the results.

The gamma spectrum was detected with a 10% coaxial Ge detector. Data acquisition was enabled only during the x-ray pulses as detected by a PIN diode. As normally used, the resulting duty cycle for the irradiation on the target was about 0.7%. The Ge detector was placed at about 37 cm distance in a plane perpendicular to the axis of the x-ray beam. Careful beam collimation, detector shielding, and use of a 3 mm. Pb + 3 mm. Cu absorber in front of the detector provided sufficiently low rates of scattered x-rays.

Thus, the dead time, resolution, and pile-up problems were insignificant even at the time of the maximum of the x-ray pulse. Portions of typical spectra are shown in Fig. 1, together with the differences obtained by subtracting raw data taken with and without x-ray irradiation under different conditions.

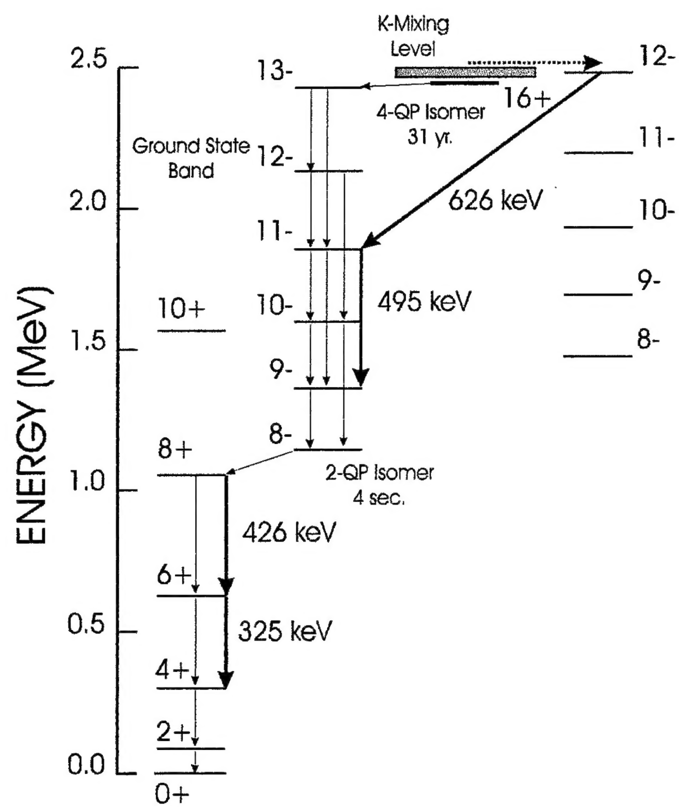

Figure 2. Energy level diagram for ^{178}Hf showing a selection of levels relevant to this experiment. The spontaneous decay of the 31-year, 16^{+} isomer is shown by the thin arrows. The thick solid arrows show components studied in this work from the particular cascade from the decay forced by the x-ray irradiation. Heavy dotted arrows are inferred transitions needed to feed those shown.

The target contained an impurity of ^{172}Hf, principally radiating the well-known decay spectrum of ^{172}Hf and its daughters at a level comparable to the intensity of the 31-year spontaneous decay of the ^{178}Hf isomer. A total of more than 100 lines were found and each was identified with one of these two nuclides or with natural background. There was no evidence of any previously unexpected nuclide. Components of the ^{172}Lu decay provided a convenient fiducial quantity against which to compare measured intensities of the emission from the isomer made with and without irradiation. The photon induced depopulation is possible for the isomeric state, but not for e-capture radioactive nuclei, such as ^{172}Hf and its daughters. The relevant energy levels of the ^{178}Hf nuclide are shown in Fig. 2. The spontaneous decay of the 31-year isomer, shown by the thin arrows is entirely through the $K^{\pi} = 8^{-}$ band followed by cascade through the ground state band, GSB beginning with the 8+ member. Because of the Pb filter neither the (2+ $\rightarrow$ 0+) line at 93.2 keV, nor the (4+ $\rightarrow$ 2+) line at 213.4 keV could be recorded with acceptable statistical accuracy.

Figure 3 shows the difference between spectra recorded in experimental series of sufficient duration to obtain about 6 x 10^4 counts in the prominent 1094 keV line of ^{172}Lu, used as a reference. In one series the target was exposed to the x-rays and in the other it was not. To acquire to within 0.3% the same fiducial counts during the exposure required for counting 546 sec. which occupied nearly 24 hrs of laboratory time. Evidence for the enhanced collection of gamma photons from some lines during the time the isomeric nuclei were exposed to the x-ray illumination is shown in Fig. 3. The sequences of lines enhanced were complex and at this time only the one for which measured components are shown by the heavy arrows in Fig. 3 has been analyzed. It begins with the (12- $\rightarrow$ 11-) line at 626.2 keV not present in the natural decay of the isomer and feeds the (11- $\rightarrow$ 9-) transition at 495.0 keV. However, the number of extra gamma photons forced through the 626.2 line is only about 1/3 of the number found in the enhancement of the 495.0 line. So there must be additional feeding from other cascades, but these do not include feeding from the 574.2 (13- $\rightarrow$ 11-) transition in the same band as found in the natural decay. That line at 574.2 keV showed no detectable enhancement when the sample was irradiated. The enhancement of the 426.4 keV line emitted from the lower energy 8+ level of the

GSB is less than seen in the feeding transitions measured in the 8^- band, but this was expected from the combination of a 2 sec. measurement interval coincident with the duration of the irradiation and the 4 sec. halflife of the 8^- bandhead delaying some emission from the lower part of the cascade past each measurement period. However, additional feeding of the GSB from other cascades not present in the normal decay of the isomer can not be excluded at this time. From Fig. 3 the enhancement can be estimated to be about 4% ± 1% of the intensity of the 495.0 keV line emitted in the natural decay of the isomer.

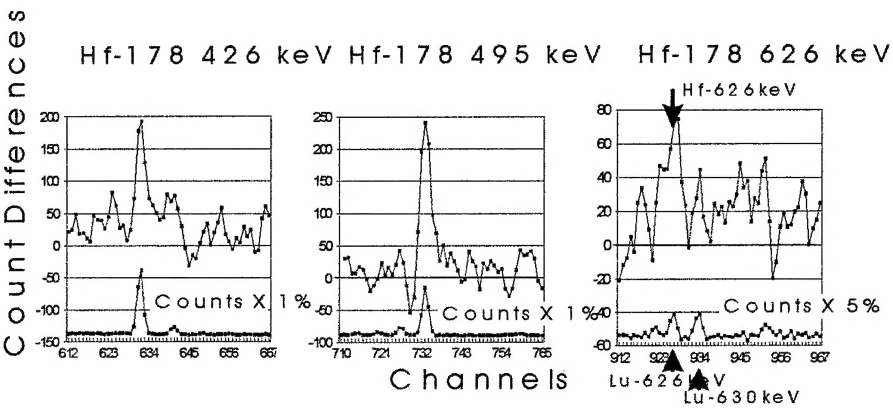

Figure 3. Plot of the differences in counts obtained in the spectrum of the ^{178}Hf from the isomeric target when it was exposed to the x-ray beam and when it was not. From left to right, the three lines correspond to the transitions ($8^+ \rightarrow 6^+$), ($11^- \rightarrow 9^-$), and ($12^- \rightarrow 11^-$). Also shown are the spectra from the unirradiated target scaled by 1% or 5% to facilitate comparison.

IV. Discussions and Conclusions

In a simple model the yield of triggering events would equal the product of the number of isomeric atoms in a target, the spectral flux density at the appropriate energy, and the unknown integrated cross section for the branch of the excitation of a K-mixing level that ends in a state other than that of the initial isomer. Since each quantity is known except for the integrated cross section for the "triggering branch," that cross section can be obtained if the transition energy is estimated. The energy for the highest intensity of the incident photons that could excite the 12^- state is about 40 keV, and this value was assumed for the cross section estimate to give an order-of-magnitude. At 40 keV an integrated cross section, $\sigma\Gamma$ is obtained:

$$\sigma\Gamma = 1 \times 10^{-21} \ \ \mathrm{cm}^2\,\mathrm{keV}, \qquad (1)$$

with an uncertainty of at least 25%. This estimate can be seen to be quite consistent with that reported [5] for ^{180}Ta, taking into account the lower triggering energy appearing in the cross section of the absorption in the present case with respect to the ^{180}Ta case. Bounds on the transition energy, E_k to the K-mixing level can be obtained from the low energy cut-off seen in the actual x-ray spectra of 20 keV and the substantial loss of intensity above 60 keV when the endpoint was changed from 90 to 70 keV. While the excitation of the decay chain starting from the 12^- level would require 40 keV, there could be other cascades originating from a different fragment of the K-mixing level. We report,

$$E_k = 40 \pm 20 \ \mathrm{keV}. \qquad (2)$$

For the case of isomeric ^{178}Hf we have demonstrated that the resonant absorption of an x-ray photon with the energy of the order of 40 keV can induce the prompt release of the 2.446 MeV stored by the isomer into freely radiating states. This is an energy gain of about 60. Only 9 mW cm^{-2} of total x-ray power in the bremsstrahlung were required at the target. Further research is needed to provide greater precision to the measurements of the transition energy to the K-mixing level needed to trigger the release of the stored isomeric energy. Such data will then facilitate a better understanding of these first evidences of the triggering of induced gamma emission

from the 31-year isomer of ^{178}Hf with very low energy x-ray photons through such large cross sections.

The results obtained from these measurements were published in Ref. [8,9].

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This report results from a contract tasking Romanian Center for Induced Gamma Emission as follows: The contractor will (a) travel to Orsay, France to participate in the data collection; (b) reduce some of the data at RC-IGE in Bucharest; © review the data and decide analytical strategies; and (d) review results and develop a seque f publications summarizing the results.

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